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Splitting of the dispersion relation of surface plasmons on a rough self-affine fractal surface

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The surface-plasmon dispersion relation for a statistically rough surface can display a splitting under specific conditions. Calculations of the dispersion-relation splitting are performed in terms of a correlation model or self-affine fractal rough surfaces with analytic form of the associated roughness spectrum, $G(\mathbf{K}) \propto (1 + aK^2\xi^2)^{-1-H}$. The dependence of the splitting on the roughness exponent H is investigated.

At an ideal flat metal surface coupling between surface plasmons and light is not permissible because momentum cannot be conserved parallel to the surface. However, as pointed out first by Stern and Ferrell,¹ the inability of a smooth surface to optically excite surface plasmons can be frustrated by allowing surface roughness to conserve momentum tangential to the surface, and thus allowing interaction with the radiation field. The knowledge of the dispersion relation of surface plasmons is of fundamental interest in surface physics as attempts are made to characterize the roughness of surfaces as well as to encounter the roughness effect on various other properties. There is a wide variety in nature of surfaces and interfaces which are described in terms of self-affine fractal scaling, for example, the nanometer scale topology of vapor deposited metal films under nonequilibrium conditions.² The effect of this kind of surface roughness can be studied in many cases quantitatively, since roughness enters through the surface height-height correlation function or its Fourier transform the roughness spectrum.^{3-8,11,19}

Up to now a quantitative analysis of the effects of surface roughness on the dispersion relation for surface plasmons has been performed in an older work for a special form of the roughness spectrum, $G(\vec{K}) \propto \delta(\vec{K} - \vec{K}_r)$, which assumes that the surface roughness has a wavelength which peaks around a given value λ_r , and corresponds to a wavy surface.^{3,5} Recently, in another study, the wave spectrum of surface plasmons has been examined for random Gaussian roughness with $G(\vec{K}) \propto e^{-aK^2}$.⁴ However, none of the previous cases correspond to rough surfaces that are grown under nonequilibrium conditions and characterized with self-affine fractal scaling. In this case, the roughness spectrum is characterized with a power-law behavior over finite length scales where, apart from the effect of a characteristic length scale,^{1,6} the degree of surface irregularity plays important role. The degree of surface irregularity is described by a roughness exponent H . Therefore, an investigation of the impact of the surface irregularity on the dispersion relation of surface plasmons is in order. In the present paper, we shall perform our calculations in terms of a simple model for the dispersion relation on a rough surface,³ since more refined treatments on random rough surfaces leave largely unchanged the qualitative result of the model under consideration.⁴

Dispersion relation. The dispersion relation for surface plasmons on a smooth solid vacuum boundary is given by $n_s(K) = \epsilon k_2 + K_1 = \epsilon(K^2 - 1)^{1/2} + (K^2 - \epsilon)^{1/2} = 0$, from which we obtain $\epsilon = -K^2(K^2 - 1)^{-1}$. At large wave vectors K , $\epsilon(\omega) \approx -1$. K is the magnitude of the surface-plasmon wave vector in units of ω/c with ω being the surface plasmon frequency. For a rough surface the dispersion relation has to be modified in such a way to include roughness effects.^{3,4,7,8} Krestschmann, Ferrell, and Ashley³ have shown that in the regime of large wave vectors K ($K > 2$) the dispersion relation can be simplified to the form

$$\frac{n_r(\vec{K})}{K} = \epsilon + 1 - \frac{\omega^2}{c^2}(\epsilon - 1)^2 \frac{I_1(\vec{K})}{\epsilon + 1} = 0, \quad (1)$$

$$I_1(\vec{K}) \approx \frac{\omega^2}{c^2} \int_{K' > 2} d^2\vec{K}' G \left[\frac{\omega}{c} |\vec{K} - \vec{K}'| \right] K K' (1 - \cos\phi)^2, \quad (2)$$

with $\cos\phi = \vec{K} \cdot \vec{K}' / KK'$. Equation (1) leads to two solutions for $\epsilon = \epsilon(\omega)$ which are given by $[\epsilon(\omega) + a_1][\epsilon(\omega) + a_2] = 0$, with

$$a_1 = \left[1 - \left[\frac{\omega^2}{c^2} I_1(\vec{K}) \right]^{1/2} \right] / \left[1 + \left[\frac{\omega^2}{c^2} I_1(\vec{K}) \right]^{1/2} \right],$$

and $a_2 = 1/a_1$. The roots a_1 and a_2 determine the separation of the peaks which is given by³

$$\Delta\epsilon = a_2 - a_1 \approx 4 \left[\frac{\omega^2}{c^2} I_1(\mathbf{K}) \right]^{1/2}.$$

The splitting takes place near the surface plasma resonance where one may observe a double peak. The physical reason for the splitting in the case of a statistically rough surface is that nearly all wave vectors of the roughness spectrum can produce interactions in between surface plasmons with different directions of the wave vectors since in the regime of large K ($K > 2$) the dispersion relation is flat (independent of magnitude and direction of the wave vector). Surface excitations with dispersion relation, which is wave-vector dependent, do not display roughness-induced splitting.

Roughness model. The type of rough surface in the static phase we shall consider here, is the so-called solid-on-solid model in which the surface is defined by a vertical height profile above a horizontal xy plane, and is

represented by a single-valued random function $z(\vec{r})$ of the in-plane positional vector $\vec{r}=(x,y)$. The difference $z(\vec{r})-z(\vec{r}')$ is assumed to be a Gaussian random variable whose distribution depends on the relative coordinates $(x'-x, y'-y)$ such that $g(\vec{R})=\langle [z(\vec{r})-z(\vec{r}')]^2 \rangle$, $\vec{R}=\vec{r}'-\vec{r}$. For an isotropic surface in x - y directions we may assume that $g(\vec{R})=AR^{2H}$ with $0<H<1$. This kind of surface roughness can be attributed to self-affine fractal surfaces as defined by Mandelbrot in terms of fractional Brownian motion.⁹ The roughness exponent H determines the surface texture of the degree of surface irregularity, and is associated with a local fractal dimension $D=3-H$.^{9,10} For $R\rightarrow\infty$, $g(\vec{R})\rightarrow\infty$, and $g(\vec{R})/R^2\rightarrow 0$ (surface asymptotically flat) which is a rather ideal case since on real surfaces $g(\vec{R})$ at large length scales may saturate to the value $2\sigma^2$. This implies the existence of an effective roughness cutoff ξ (correlation length) such that for $R\ll\xi$, $g(\vec{R})\propto R^{2H}$ and for $R\gg\xi$, $g(\vec{R})\approx 2\sigma^2$.^{11,12} The parameter $\sigma=\langle z(\vec{0})^2 \rangle^{1/2}$ is the rms saturated surface roughness. The height-height correlation $C(\vec{R})=\langle z(\vec{R})z(\vec{0}) \rangle$ is related to $g(\vec{R})$ by $g(\vec{R})=2\sigma^2-2C(\vec{R})$.

We define the Fourier transform of $z(\vec{R})$ by

$$z(\vec{K})=(2\pi)^{-2}\int z(\vec{R})e^{-i\vec{K}\cdot\vec{R}}d\vec{R},$$

and the height-height correlation by

$$C(\vec{R})=A^{-1}\int \langle z(\vec{P})z(\vec{P}+\vec{R}) \rangle d\vec{P}.$$

The roughness spectrum is given by

$$\langle |z(\vec{K})|^2 \rangle = \frac{A}{(2\pi)^4} \int C(\vec{R})e^{-i\vec{K}\cdot\vec{R}}d\vec{R} \quad (3)$$

with A being the macroscopic surface area. The function $G(\vec{K})$ is related to $\langle |z(\vec{K})|^2 \rangle$ by

$$G(\vec{K}) = \frac{(2\pi)^4}{A} \langle |z(\vec{K})|^2 \rangle,$$

and is normalized such that $\int G(\vec{K})d\vec{K}=\sigma^2$. There is a specific class correlation functions for self-affine fractals,⁶ called K -correlations, with an analytic form of roughness spectrum,

$$\langle |z(\vec{K})|^2 \rangle = \frac{A}{(2\pi)^5} \frac{\sigma^2 \xi^2}{(1+aK^2\xi^2)^{1+H}}. \quad (4)$$

The parameter a is given by

$$a=1/2H[1-(1+a\pi^2\xi^2/a_0^2)^{-H}]$$

with a_0 being the atomic spacing of typical size $a_0\sim 3\text{ \AA}$. In the limit $H\rightarrow 0$, Eq. (4) lead to correlations related to logarithmic roughness which are encountered in various roughening transition systems,^{13,14} where the parameter a in this case takes the form, $a=\frac{1}{2}\ln(1+a\pi^2\xi^2/a_0^2)$. The correlation functions associated with Eq. (4) have the general form⁶

$$C(\vec{R}) = \frac{\sigma^2}{a\Gamma(1+H)} \left[\frac{R}{2\xi\sqrt{a}} \right]^H K_H \left[\frac{R}{\xi\sqrt{a}} \right]. \quad (5)$$

Since the history related to the dispersion relation of surface plasmons is valid for slight rough surfaces,³ we shall

restrict our study to the case of $\sigma\ll\xi$.

Selected numerical results—discussion. The plot of the response function $|K/n_r|^2$ [given by Eq. (1)] versus $\text{Re}(\epsilon)$ possesses maxima (peaks) at which their positions give the values of $\epsilon(\omega)$, at which surface-plasmon excitation is possible. The plots of the response function have been performed with $\text{Im}(\epsilon)=0.1$, a surface-plasmon wave vector $K(=K_p)=10.0$ (in units of ω/c), and a wavelength of electromagnetic waves in vacuum $\lambda=4000.0\text{ \AA}$, which implies $\lambda_p=400\text{ \AA}$. Therefore, the present calculations correspond to $\lambda_p=400\text{ \AA}$, as well as the other parameters [ω/c , and $\text{Im}(\epsilon)$] are the same as for potassium (Ref. 3). In our study, we shall focus mainly on the dependence of the dispersion relation splitting on the parameters H and ξ . The effect of σ with respect to ξ has been investigated in previous studies.^{3,4} In Fig. 1 we plot the response function for various values of the roughness exponent H in the regime $0.0\leq H\leq 0.8$ for fixed values of ξ and σ , and it becomes clear that the higher the degree of irregularity (smaller H) the larger is the splitting. Alternatively, this can be explained from the fact that as H is decreased with fixed σ , the amplitude of the correlation function $g(\vec{R})$ [and equivalently $G(\vec{K})$] increases. In Fig. 2 we plot the response function for various values of the correlation length ξ with H and σ fixed. As the correlation length increases the splitting becomes weaker, and behavior observed for a smooth surface boundary is approached. This can be explained from the fact that as ξ increases with fixed σ the ratio σ/ξ decreases, resulting in an increasing surface smoothness.

However, in order to gauge which parameter between H and ξ has the dominant effect on the splitting of the dispersion relation of surface plasmons, we calculate the response function for the case of logarithmic roughness ($H=0$) since for a given value of ξ the splitting is maximum (Fig. 1), over a wide range of correlation lengths

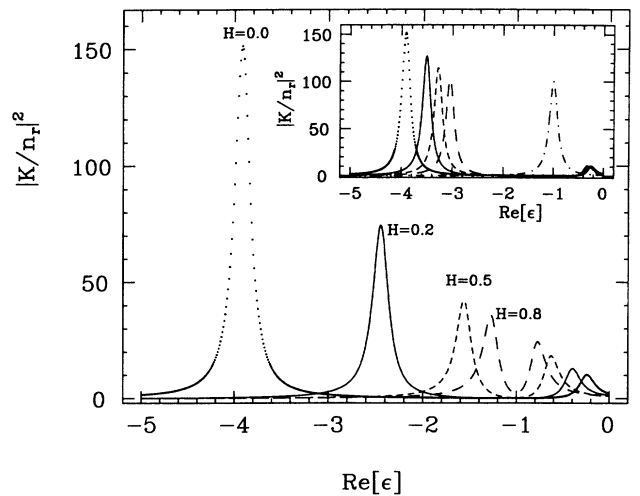


FIG. 1. Schematics of the response function for $a_0=3\text{ \AA}$; $\sigma=20.0\text{ \AA}$; $\xi=400.0\text{ \AA}$, for various values of the roughness exponent H . The inset shows the splitting for the case of logarithmic roughness, $\sigma=20.0\text{ \AA}$; $H=0$; $\xi=400.0\text{ \AA}$: dots; $\xi=800.0\text{ \AA}$: solid line; $\xi=2000.0\text{ \AA}$: small dashes; $\xi=5000.0\text{ \AA}$: large dashes; smooth surface boundary: dot dashes.

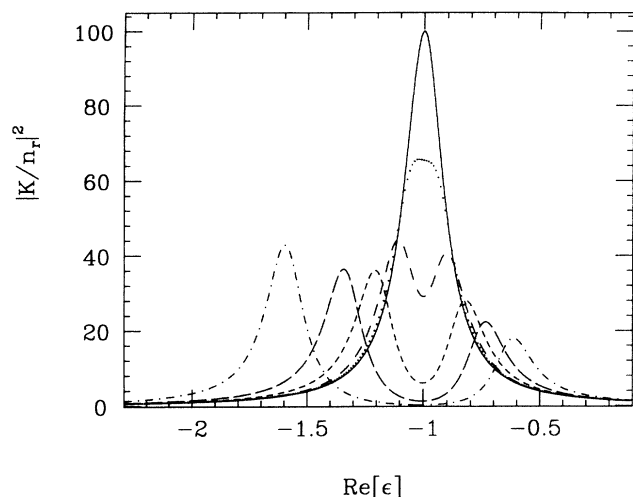


FIG. 2. Schematics of the response function for $a_0 = 3$ Å; $\sigma = 20.0$ Å; $H = 0.7$, as a function of the correlation length ξ , smooth surface boundary: solid line; $\xi = 200.0$ Å: dot dashes; $\xi = 400.0$ Å: large dashes; $\xi = 800.0$ Å: small dashes; $\xi = 2000.0$ Å: dashes; $\xi = 5000.0$ Å: dots.

$400.0 \leq \xi \leq 5000.0$ Å, inset Fig. 1. The schematics show that even for large correlation lengths, the splitting remains significantly large. Therefore, we can conclude that the dominant factor in the splitting of the dispersion relation for surface plasmons on self-affine rough surfaces is the degree of surface irregularity which is expressed through the roughness exponent H . Furthermore, our calculations reveal an interplay between correlation length ξ and local fractal dimension $D = 3 - H$, where an

incremental behavior of both parameters has an opposite result regarding the splitting of the dispersion relation (peak separation). Furthermore, it should be pointed out that with decreasing H (which results in larger splitting), the height of the first peak is increased while the height of the second peak is decreased significantly. This point is of crucial importance, since it is relevant to the experimental detection of the splitting of the dispersion relation.¹⁵

In conclusion, the aim of this report was to correlate known information concerning the dispersion relation of surface plasmons for a particular model of surface roughness, the self-affine fractal roughness, which is widely observed to develop during thin-film fabrication under nonequilibrium conditions. It came out through this study in a qualitative level that the local fractal dimension $D = 3 - H$ plays a dominant role on the splitting of the dispersion relation for a wide range of correlation lengths ξ which their magnitude has been observed in various surface roughness studies.^{6,11,12} The splitting of the dispersion relation has been experimentally observable, and possibly can be used as an alternative probe for surface roughness studies in terms for example of ellipsometric methods in cases where other techniques are not applicable [x rays, scanning tunneling microscopy (STM), etc].¹⁵ Furthermore, the observation of surface plasmons experimentally also with the STM can open new perspectives in this field,¹⁶ as well as to various other related fields,¹⁷ since Eqs. (4) and (5) offer a general way to model surface roughness observed in nature.

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